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CHICAGO UNIV IL JAMES FRANCK INST

F/G 20/5

GAS SURFACE DYNAMICS, HIGH POWER LASER-SOLID INTERACTIONS-(U)

OCT 80 L WHARTON

AFOSR-77-3186

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AFOSR-TR-80-1114

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

1. REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM	
18	AFOSR-TR-80-1114	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
A. TITLE (and Subtitle)		B. CONTRACT OR GRANT NUMBER(s)	
6 Gas Surface Dynamics, High Power Laser-Solid Interactions.		15 AFOSR - 77 - 3186	
C. AUTHOR(s)		D. PERFORMING ORG. REPORT NUMBER	
10 Lennard Wharton		Final Report.	
E. PERFORMING ORGANIZATION NAME AND ADDRESS		F. DATE OF REPORT (and Period Covered)	
The James Franck Institute and The Department of Chemistry, The University of Chicago, 5801 S. Ellis. Chicago, IL 60637		16 January 1978 - Dec. 1979	
G. CONTROLLING OFFICE NAME AND ADDRESS		H. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS	
AFOSR/NC Bolling AFB, DC 20332		61102F 2303/A2	
I. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		J. REPORT DATE	
12 11		11 6 October 1980	
K. SECURITY CLASS. (of this report)		L. NUMBER OF PAGES	
Unclassified		9	
M. DISTRIBUTION STATEMENT (of this Report)		N. DECLASSIFICATION/DOWNGRADING SCHEDULE	
Approved for public release; distribution unlimited.			
O. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)			
P. SUPPLEMENTARY NOTES			
80 11 06 183			
Q. KEY WORDS (Continue on reverse side if necessary and identify by block number)			
Surface Reactions Laser Laser-surface interactions Energy exchange Surface Science		Surface chemistry Molecular beams	
R. ABSTRACT (Continue on reverse side if necessary and identify by block number)			
This work is a continuation of studies of fundamental aspects of the dynamics of chemical and physical processes on surfaces. The use of pulsed molecular beams and pulsed laser energy beams to initiate and probe energy exchange and chemical reactions that occur between gases and metal surfaces has been made in experimental studies. The product species and translational kinetic energies are determined with the aid of a mass spectrometer capable of determining the time of arrival of these species. These studies are			

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relevant to the effects of high power laser beams on surfaces. Studies included the following gases: NO_2 , O_2 , CO , H_2 , Ar, He, Ne, Xe, and the following surfaces W, Pt, Pt(111).

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DDC TAB	<input type="checkbox"/>
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Availability Codes	
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I. Research Report

FINAL

A. RESEARCH OBJECTIVES

This is a study of specialized aspects of fundamental processes of gas-surface dynamics. The studies include a range of absorbate-substrate chemical and physical combinations. Particular emphasis is directed toward understanding the processes of desorption at especially high rates, even though the experiments are conducted under ultra-high vacuum conditions. An important technique under development is the extremely rapid temperature jump introduced by laser radiation heating a metallic substrate. The ensuing temperature ramp serves as a shutter for observing the velocity distribution by time-of-flight methods of the desorbing species. Further studies of gas-surface dynamics are conducted with the aid of a doubly differentially pumped rotatable mass spectrometer detector and highly differentially pumped supersonic and thermal dosing beams. These studies are relevant to effects of high power laser beams on surfaces, in particular the initial products of such situations. The instrumentation and techniques developed here may prove useful diagnostics to understand the causes of susceptibility to laser damage and remedies for it. The studies give fundamental information on processes and reactions that occur at high speeds and high temperatures involving gases and solids. These include evaporation, chemical reactions, and energy exchange. Related studies have included measurement of the velocity and angular distributions of atoms and molecules involved in physical and chemical gas-surface processes, in which the initiation and control comes from high speed mechanical chopping of molecular beams. These studies are complementary to the laser studies because they give a more complete picture of a particular process.

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B. RESULTS

The final results are expected in detail in the 12 publications in print and 5 manuscripts in preparation. However it is worthwhile to highlight the findings here.

(1) The apparatus for the investigation of gas-surface dynamics was entirely completed and worked up to expectations.

(2) Two studies were performed using gas-surface laser desorption, an earlier one desorbing H_2 from W, and a later one, with the improved laser, desorbing H_2 from Pt(111). The later Pt studies have extended our knowledge of the phenomena of H_2 desorption from Pt to such a high temperature and surface coverage that a much more reliable measure of the activation energy of desorption and the pre-exponential frequency factor of this process was determined than could otherwise be obtained under the controlled conditions of UHV, single crystal surface.

(3) The accommodation processes of noble gases and unreacted molecules such as O_2 and N_2 on clean metal surfaces were shown to involve accommodation coefficients that are independent of beam energy and surface temperature over wide ranges. The nature of the direct inelastic scattering channel was determined. The values of these translational accommodation coefficients have been measured, and they have been the subject of considerable theoretical modeling of gas-surface energy exchange, stimulated by the accurate and comprehensive measurements that have been reported by this laboratory.

(4) The study of the oxidation of CO on Pt(111) has revealed that this mechanism involves an activated complex whose energy of activation appears in the translational energy of the products.

- (5) Rich patterns of diffraction of He from H on Pt(111) have determined the exact geometry of the H overlayer on Pt(111). The H is bound in the 3-fold site on the Pt(111) in a single domain. The amplitude of the repulsion potential of this surface has been determined by modeling the He scattering with a 2-dimensional repulsion wall model.
- (6) The nature of the mechanisms of surface reaction of O_2 and N_2 on W were determined by analysis of the energy and angle of the unreacted O_2 and N_2 molecules. O_2 reacts via direct chemisorption, while N_2 reacts via a molecular precursor mechanism. The efficiency of which diminishes as the surface temperature is raised through re-evaporation and direct inelastic scattering of the N_2 .

C. ACKNOWLEDGMENT

The Principal Investigator wishes to thank the AFOSR for its helpful support and interest in his research investigation over his years at the University of Chicago. He acknowledges vital support given when most needed, under conditions when rare faith and imagination were required of the granting agency.

II. PUBLICATIONS

1. Energy Accommodation and Reactivity of O_2 on Tungsten (with Daniel Auerbach, Charles Becker and James Cowin), Appl. Phys. 14, 141 (1977).
2. Mechanism and Speed of Initial Step of Oxygen Chemisorption - O_2 on W (with Daniel Auerbach, Charles Becker and James Cowin), Appl. Phys. 14, 411 (1977).
3. CO_2 Product Velocity Distributions for CO Oxidation on Platinum (with C. A. Becker, J. P. Cowin and D. J. Auerbach), J. Chem. Phys. 67, 3394 (1977).
4. Absence of Translational Energy Accommodation of O_2 on Clean and Oxidized Tungsten, Specularly and Diffusely Scattered (with Daniel Auerbach, Charles Becker and James Cowin) Book of Abstracts (VI International Symposium on Molecular Beams, Noordwijkerhout, The Netherlands, 1977) Part II, p. 192.
5. Intermolecular Potentials and Energy Exchange Processes Between Alkali Halides and Other Gases as Determined by Scattering Experiments. Chapter in a book tentatively entitled Alkali Halide Vapors, edited by Paul Davidovits and D. L. McFadden (Academic Press, N.Y., 1978).
6. Measurement of Fast Desorption Kinetics of D_2 from Tungsten by Laser Induced Thermal Desorption (with J. P. Cowin, D. J. Auerbach and C. Becker), Surf. Sci. 78 (1978) 545.
7. Chemical Dynamics Investigated by Laser Probes, L. Wharton, D. Auerbach, D. Levy, R. Smalley, in Advances in Laser Chemistry, A. H. Zewail, ed., Springer Series in Chemical Physics (Springer, Berlin, Heidelberg, New York, 1978).
8. UHV Application of Spring Loaded Teflon Seals, Daniel J. Auerbach, Charles A. Becker, James P. Cowin and Lennard Wharton. Rev. Sci. Instrum. 49, 1518 (1978).
9. Direct Measurement of Velocity Distributions in Argon Beam-Tungsten Surface Scattering, K. C. Janda, J. E. Hurst, C. A. Becker, J. P. Cowin, Daniel J. Auerbach, L. Wharton, J. Chem. Phys. (in press).
10. Energy Accommodation and Condensation of Argon and Nitrogen on Tungsten, C. A. Becker, D. J. Auerbach, J. Cowin, K. Janda, L. Wharton and J. Hurst, Rarefield Gas Dynamics, 11th Symposium edited by R. Campayne (CEA, Paris, 1979) pp. 1427-1432.
11. Direct Inelastic and Trapping-Desorption Scattering of N_2 from Polycrystalline W; Elementary Steps in the Chemisorption of Nitrogen, K. C. Janda, J. E. Hurst, C. A. Becker, J. P. Cowin, L. Wharton, and D. J. Auerbach. Surf. Sci. 93, 270 (1980).
12. Observation of Direct Inelastic Scattering in the Presence of Trapping-Desorption Scattering: Xe on Pt(111). J. E. Hurst, C. A. Becker, J. P. Cowin, K. C. Janda, L. Wharton and D. J. Auerbach. Phys. Rev. Lett. 43 1175 (1979).

13. Oxidation of CO on Pt(111) (in preparation) C. Becker.
14. He diffraction study of Surface structure of H on Pt(111) (in preparation) J. Lee.
15. Ar, Xe Energy Accommodation on Pt(111) (in preparation) J. Hurst.
16. HD Exchanger Reaction on Pt(111) Studied by Laser Induced Desorption and Modulated Molecular Beams. (in preparation) J. Cowin.
17. CO Desorption and Adsorption on Pt(111) writer D. Winicur, J. Hurst, C. Becker, to be submitted Surf. Sci.

III. PERSONNEL

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James Cowin

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Charles Becker, PhD June 1979

(2) N.S.F. Postdoctoral Fellow

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IBM Research Laboratories
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**Professor Kenneth Janda
California Institute of Technology
Pasadena, California**

(4) Visiting Scientist Jan - June 1978

**Professor Daniel Winicur
Notre Dame University
South Bend, Indiana**

IV. INVITED TALKS, L. WHARTON

Penn State	December, 1977
Bell Labs, Murray Hill	February, 1978
University of Pennsylvania	February, 1978
Notre Dame	March, 1978
Am. Chem. Soc., Anaheim, Ca.	March, 1978
Cal Just of Technology	March, 1978
University of Texas, Austin	April, 1978
Purdue University	April, 1978
ONR Surface Sciences, IBM Yorktown Heights, NY	April, 1978
Brookhaven National Laboratory	June, 1978
Asilomar Chemical Dynamics Conference	June, 1978
Rarefield Gas Dynamics Carmes, France	July, 1978
Spectral Line Broadening Conference Winsor, Ontario, Canada	August, 1978
American Vacuum Society Cornell University Ithaca, New York	November, 1978
Laser Chemistry Conference Ein Bokek, Israel	December, 1978
University of Illinois Urbana, Illinois	April, 1979
University of Indiana Bloomington, Indiana	April, 1979
ONR Surface Science U. of Minnesota Minneapolis, Minnesota	May, 1979
Molecular Beams Conference Trento, Italy	May, 1979
Les Houches Summer School Chamouix, France	June, 1979

IV INVITED TALKS, L. WHARTON (continued)

International Conferences on
Vander Waals Molecules, Quebec

August, 1979

Gordon Conference on Surface Science
Plymouth, New Hampshire

August, 1979

American Vacuum Society
New York City

October, 1979